U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE  TRANSMITTAL LETTER TO THE UNITED STATES							
DÉSIGNATED/ELECTED OFFICE (DO/EO/US)							
CONCERNING A FILING UNDER 35 U.S.C. 371							
ATTORNEY DOCKET NUMBER	U.S. APPLICATION NO (IF NOWN SEE & RR 15)						
IN-5554 INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED					
PCT/EP00/07890	14. August 2000 (14.08.2000	16. August 1999 (16.08.1999)					
TITLE OF INVENTION: OLIGOMERS AND POLYMERS ON THE BASIS OF DIETHYL-OCTANEDIOLS, METHOD FOR							
PRODUCING SAME AND THE USE THEREOF							
APPLICATION(S) FOR DO/EO/US: Hubert BAUMGART, Heinz-Peter RINK, and Detlef KRATZ							
Applicant herewith submits to the Uni	ited States Designated/Elected Office (DO/	EO/US) the following items and other					
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:							
1. X This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.							
2. This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.							
3. This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371 (b) and PCT Articles 22 and 39(1).							
examination until the expiration and for Internation	tion of the applicable time limit set in 35 U.S. ational Preliminary Examination was made by	o. or it to j and FOT Anticles 22 and 39(1). The 19th month from the earliest claimed					
	Application as filed (35 U.S.C. 371(c)(2)) erewith (required only if not transmittal by the	International Bureau).					
b. x have been transm	itted by the International Bureau.						
c. is not required, as	the application was filed in the United States anal Application into English (35 U.S.C. 371(0						
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a are transmitted he	the International Application under PCT Artic erewith (required only if not transmitted by the						
) "	nitted by the International Bureau.						
c. have not been ma	ede; however, the time limit for making such and and will not be made	mendments has NOT expired.					
8. A translation of the amendm	nents to the claims under PCT Article 19 (35	J.S.C. 371(c)(3)).					
b. nave been transmore. c. have not been may have not been transmore.  The state of the	e inventor(s) (35 U.S.C. 371(c)(4)).						
10. A translation of the annex to the International Preliminary Examination Report under PCT Article 36							
Items 11. to 16. below concern other document(s) or information included:							
11. X An Information Disclosure Statement under 37 CFR 1.97 and 1.98.							
12. An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included							
13. X A FIRST preliminary amendment. A SECOND or SUBSEQUENT preliminary amendment.							
14. A substitute specification.							
15. A Change of power of attorn	ney and/or address letter.						
16. X Other items or information:							
A copy of the cover sheet from the PCT Published Application							
A copy of the cover sheet from the Priority Document							
	ondence is being deposited with the United States <b>EH468390720US</b> addressed to the Commissioner						
Post Office to Addressee" Mailing Label No. <u>EH468390720US</u> addressed to the Commissioner for Patents, Washington, D.C. 20231 on February 13, 2002.							
may. Ser							
Mariorie Ellis							

U.S. APPLICATION NO. (If known see 37 C.F.B. 1.50 INTERNATIONAL APPLICATION NO.		ATTORNEY'S DOCKET NUMBER				
10/049656   PCT/EP00/07890		IN-5554				
17. X The following fees are submitted			CALCULATIONS PTO USE ONLY			
		7 CFR 1.492(a)(1)-(5	o)): n fee (37 CFR 1.482)			
			(a)(2)) paid to USPTO			
and Inter	national Sear	ch Report not prepare	ed by the EPO or JPO	\$970.00		
			37 CFR 1.482) not paid to U	SPTO		
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International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4)						
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<u> </u>		-	n or declaration later than	20 30		
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Claims		Number Filed	Number Extra	Rate		
Total	Claims	19 - 20 =	0	X \$18.00	\$	
ndepende		01 - 03 =	0	X \$80.00	\$	
Multiple de	ependent claims	s(s) (if applicable)		+ \$270.00	\$	
			TOTAL OF ABOVE		\$890.00	
Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).			\$			
SUBTOTAL =			\$890.00			
Processing fee of \$130.00 for furnishing the English translation later the [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492(f)). +			\$			
TOTAL NATIONAL FEE =			\$890.00			
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +			\$ 40.00			
11010 11010 11010			TOTAL F	EES ENCLOSED =	\$930.00	
	•				Amount to be: refunded	\$
					Charged	\$930.00
a. A check in the amount of \$ to cover the above fees is enclosed.						
b. X Please charge my Deposit Account No. <u>23-3425</u> in the amount of <u>\$930.00</u> to cover the above fees A triplicate copy of this sheet is enclosed.						
c. X The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>23-3425</u> . A triplicate copy of this sheet is enclosed.						
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PATENT

(Practitioner's Docket No. IN-5554)

### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of

Hubert BAUMGART Heinz-Peter RINK Detlef KRATZ

Serial No.: This application is a National Phase of Patent Application PCT/EP00/07890 filed 14. August 2000.

Filed: February 13, 2002

For: OLIGOMERS AND POLYMERS ON THE BASIS OF DIETHYL-OCTANEDIOLS, METHOD FOR PRODUCING SAME AND THE USE THEREOF Group Art Unit: Not Assigned

Examiner: Not Assigned

I hereby certify that the attached correspondence is being deposited with the United States Postal Service in an envelope as "Express Mail Post Office to Addressee" Mailing Label No. <u>EH468390720US</u> addressed to Commissioner for Patents, Washington, D.C. 20231 on February 13, 2002.

Marjorie Ellis

## PRELIMINARY AMENDMENT UNDER 37 CFR § 1.115

Commissioner for Patents

Washington, D.C. 20231

Dear Sir:

This preliminary amendment is submitted with the application for entry into the U.S. National Phase under Chapter II. This application is based on PCT/EP00/07890 filed on 14 August 2000.

In connection with the filing of this National Phase application, please make the following preliminary amendments.

### IN THE SPECIFICATION

After the title, please insert --This application is a National Phase Application of Patent Application **PCT/EP00/07890** filed on 14 August 2000--

#### IN THE CLAIMS:

Please cancel claims 1-20.

Please add claims 21-39.

- 21. (New) A composition comprising a reaction product of a positionally isomeric diethyloctanediol monomer, wherein the reaction product is at least one of an oligomer and a polymer.
- 22. (New) The composition of claim 21, wherein the positionally isomeric diethyloctanediol is a 2,3-, 2,4-, 2,5-, 2,6-, 2,7-, 3,4-, 3,5-, 3,6- or 4,5- diethyloctanediol.
- 23. (New) The composition of claim 22, wherein the positionally isomeric diethyloctanediol is a 2,4-diethyloctanediol.
- 24. (New) The composition of claim 21, wherein the positionally isomeric diethyloctanediol is a diethyloctane-1,2-, -1,3-, -1,4-, -1,5-, -1,6-, -1,7-, -1,8-, -2,3-, -2,4-, -2,5-, -2,6-, -2,7-, -2,8-, -3,4-, -3,5-, -3,6-, -3,7-, -3,8-, -4,5-, -4,6-, -4,7-, -4,8-, -5,6-, -5,7-, -5,8-, -6,7-, -6,8- or -7,8-diol.
- 25. (New) The composition of claim 24, wherein the positionally isomeric diethyloctanediol is a diethyloctane-1,5-diol.
- 26. (New) The composition of claim 25, wherein the positionally isomeric diethyloctanediol is a 2,4-diethyloctane-1,5-diol.
- 27. (New) The composition of claim 21, wherein the reaction product is at least one of a polyaddition reaction product and a polycondensation reaction product.
- 28. (New) The composition of claim 27, wherein at least one of the polyaddition reaction product and the polycondensation reaction product comprise at least one of a polyether, a polyester, a polycarbonate, a polyurethane, a polyurea, a polyamide, a polyimide, an ether copolymer, an ester copolymer, a carbonate copolymer, a urethane copolymer, a urea copolymer, an amide copolymer, and an imide copolymer.

- 29. (New) The composition of claim 28, wherein at least one of the polyaddition reaction product and the polycondensation reaction product comprise at least one of a polyester, a polyurethane, a polyester-co-polyether, a polyester-co-polycarbonate, a polyester-co-polyurethane, a polyester-co-polyamide, a polyester-co-polyurea, and a polyester-co-polyimide.
- 30. (New) The composition of claim 21, wherein the reaction product has a structure that is at least one of linear, branched, block, comb, random, core/shell, and the form of crosslinked microparticles.
- 31. (New) The composition of claim 21, wherein the reaction product contains at least one of
  - functional groups that undergo crosslinking reactions with complementary functional groups present in at least one of the reaction products themselves and in separate compounds, and
  - ii) functional groups, which on exposure to actinic radiation, react with at least one of one another and with other groups.
- 32. (New) The composition of claim 21, wherein the reaction products are grafted with olefinically unsaturated monomers.
- 33. (New) The composition of claim 21, wherein the composition is one of a molding compound, an adhesive, a coating material, and a paint.
- 34. (New) The composition of claim 21, wherein the composition is one of a molding, a film, a fiber, an adhesive film, a decorative coating, a protective coating, and a decorative and protective coating.
- 35. (New) A method comprising applying the composition of claim 21 to a substrate.
- 36. (New) The substrate prepared by the method of claim 35.

- 37. (New) The method of claim 35, wherein the substrate is one of a motor vehicle body, an industrial component, an electrical component, a coil, a package, and furniture.
- 38. (New) The method of claim 35, wherein at least one of the following:
  - a. the positionally isomeric diethyloctanediol is a 2,3-, 2,4-, 2,5-, 2,6-, 2,7-, 3,4-, 3,5-, 3,6- or 4,5-diethyloctanediol;
  - b. the positionally isomeric diethyloctanediol is a diethyloctane-1,2-, -1,3-, -1,4-, -1,5-, -1,6-, -1,7-, -1,8-, -2,3-, -2,4-, -2,5-, -2,6-, -2,7-, -2,8-, -3,4-, -3,5-, -3,6-, -3,7-, -3,8-, -4,5-, -4,6-, -4,7-, -4,8-, -5,6-, -5,7-, -5,8-, -6,7-, -6,8- or -7,8-diol;
  - c. the positionally isomeric diethyloctanediol is a 2,4-diethyloctane-1,5-diol;
  - d. the reaction product is at least one of a polyaddition reaction product and a polycondensation reaction product;
  - e. the reaction product is at least one of a polyether, a polyester, a polycarbonate, a polyurethane, a polyurea, a polyamide, a polyimide, an ether copolymer, an ester copolymer, a carbonate copolymer, a urethane copolymer, a urea copolymer, an amide copolymer, an imide copolymer, a polyester-copolyether, a polyester-co-polycarbonate, a polyester-co-polyurethane, a polyester-co-polyamide, a polyester-co-polyurea, and a polyester-copolyimide;
  - f. the reaction product has a structure that is at least one of linear, branched, block, comb, random, core/shell, and the form of crosslinked microparticles;
  - g. the reaction product contains at least one of
    - functional groups that undergo crosslinking reactions with complementary functional groups present in at least one of the reaction products themselves and in separate compounds, and
    - ii) functional groups, which on exposure to actinic radiation, react with at least one of one another and with other groups;
  - h. the reaction products are grafted with olefinically unsaturated monomers;
  - i. the composition is one of a molding compound, an adhesive, a coating material, and a paint.
- 39. (New) The substrate prepared by the method of claim 38.

#### **REMARKS**

Upon entry of the present amendment claims 21-39 are pending in the application. New claims 21-39 add no new matter, as these claims contain subject matter deleted from the canceled claims. There are one (1)independent claim and a total of nineteen (19) claims pending in the application. Applicants respectfully request entry of the preliminary amendment.

Respectfully Submitted,

Michael F. Morgan Registration No. 42,906

Date: February 13, 2002 BASF Corporation 26701 Telegraph Road Southfield, Michigan 48034-2442 (248)-948-2355 Customer No. 26922

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PCT/EP00/07890

# Oligomers and polymers based on diethyloctanediols, their preparation and use

The present invention relates to novel oligomers and polymers containing positionally isomeric diethyloctanediols as monomer units. The present invention further relates to the use of the novel oligomers and polymers for preparing molding compounds, adhesives, and coating materials, especially paints, and their successor products, i.e., the moldings, films, fibers, adhesive films, and coatings, especially paint systems.

order to flexibilize polyesters or copolymers In 15 containing polyester units, and moldings, adhesive films, and coatings, especially paint systems, produced using them, it is customary to employ the C6 units hexane-1,6-diol and/or adipic acid. Furthermore, polyesters and polyester-co-polyurethanes containing in 20 cocondensed form 2-methyl-1,8-octanediol and also, if desired, nonane-1,9-diol as C9 monomer units are known from numerous Japanese patent applications such as, for example, JP-A-63182330, JP-A-01215876, JP-A-01242674, JP-A-02264082, JP-A-06329751, JP-A-0493316, 02264082, JP-A-02147238 or JP-A-02041379, the European 25 patent EP-A-0 562 577 or from the British patent GB-A-2248844. Furthermore, the Japanese patent JP-A-09272731

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reveals a polyester containing 2,7-dimethyl-1,8-octanediol as a C10 monomer unit.

These oligomers and polymers have performance properties which are already good; however, in the variation of their profile of properties, for example, the further flexibilization, an upper limit is reached the crossing of which is accompanied by manifest disadvantages, such as a reduction in hardness. If, therefore, there is the desire to further vary and improve their performance properties in order that they can be put to new end uses and/or offer new advantages in their existing end uses, it will be necessary to provide new monomer units and new oligomers polymers.

It is an object of the present invention, accordingly, to provide oligomers and polymers as alternatives to the existing ones. The new polymers and oligomers are to retain the advantageous properties of the known oligomers and polymers while additionally offering new possibilities for varying the profile of properties, advantageously broadly, so that they are also suitable for new end uses.

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The invention accordingly provides the novel oligomers and polymers which contain at least one positionally isomeric diethyloctanediol as monomer unit.

In the text below, these novel oligomers and polymers are referred to as "inventive oligomers and polymers".

The invention also provides the novel molding compounds, adhesives, and coating materials which comprise at least one inventive oligomer and/or polymer. In the text below, these novel molding compounds, adhesives, and coating materials referred to as "inventive molding compounds, adhesives, 10 and coating materials".

The invention additionally provides novel moldings, films, and fibers which can be produced from the molding compounds of the invention. In the text below they are referred to as "inventive moldings, films, and fibers".

The invention further provides novel adhesive films which can be produced from the inventive adhesives and are referred to below as "inventive adhesive films".

The invention not least provides novel coatings which can be produced from the inventive coating materials and are referred to below as "inventive coatings".

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Furthermore, the invention provides novel substrates which have at least one inventive adhesive film and/or at least one inventive coating and are referred to below as "inventive substrates".

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For the purposes of the present invention, oligomers are compounds containing from 2 to 15 repeating monomer units. For further details, refer to Römpp Lexikon Lacke und Druckfarben, Georg Thieme Verlag, Stuttgart, New York, 1998, page 425, "Oligomers".

By contrast, for the purposes of the present invention, polymers are compounds which contain more than 10, in particular more than 15, repeating monomer units. For further details, refer to Römpp Lexikon Lacke und Druckfarben, Georg Thieme Verlag, Stuttgart, New York, 1998, page 464, "Polymers".

The essential constituent of the inventive oligomers

15 and polymers is at least one positionally isomeric diethyloctanediol.

The positionally isomeric diethyloctanediols for inventive use contain a linear C8 carbon chain.

With regard to the two ethyl groups, the C8 carbon chain has the following substitution pattern: 2,3, 2,4, 2,5, 2,6, 2,7, 3,4, 3,5, 3,6 or 4,5. In accordance with the invention it is of advantage if the two ethyl groups are in positions 2 and 4, i.e., if the compounds are 2,4-diethyloctanediols.

With regard to the two hydroxyl groups, the C8 carbon chain has the following substitution pattern: 1,2, 1,3,

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1,4, 1,5, 1,6, 1,7, 1,8, 2,3, 2,4, 2,5, 2,6, 2,7, 2,8, 3,4, 3,5, 3,6, 3,7, 3,8, 4,5, 4,6, 4,8, 5,6, 5,7, 5,8, 6,7, 6,8 or 7,8. In accordance with the invention it is of advantage if the two hydroxyl groups are in positions 1 and 5, i.e., if the compounds are diethyloctane-1,5-diols.

The two substitution patterns are combined with one another in any desired way; i.e., the diethyloctanediols for inventive use comprise

- 2,3-diethyloctane-1,2-, -1,3-, -1,4-, -1,5-, -1,6-, -1,7-, -1,8-, -2,3-, -2,4-, -2,5-, -2,6-, -2,7-, -2,8-, -3,4-, -3,5-, -3,6-, -3,7-, -3,8-, -4,5-, -4,6-, -4,7-, -4,8-, -5,6-, -5,7-, -5,8-, -6,7-, -6,8- or -7,8-diol,
- 2,4-diethyloctane-1,2-, -1,3-, -1,4-, -1,5-, -1,6-, -1,7-, -1,8-, -2,3-, -2,4-, -2,5-, -2,6-, -2,7-, -2,8-, -3,4-, -3,5-, -3,6-, -3,7-, -3,8-, -4,5-, -4,6-, -4,7-, -4,8-, -5,6-, -5,7-, -5,8-, -6,7-, -6,8- or -7,8-diol,
- 2,5-diethyloctane-1,2-, -1,3-, -1,4-, -1,5-, -1,6-, -1,7-, -1,8-, -2,3-, -2,4-, -2,5-, -2,6-, -2,7-, -2,8-, -3,4-, -3,5-, -3,6-, -3,7-, -3,8-, -4,5-, -4,6-, -4,7-, -4,8-, -5,6-, -5,7-, -5,8-, -6,7-, -6,8- or -7,8-diol,
  - 2,6-diethyloctane-1,2-, -1,3-, -1,4-, -1,5-, -1,6-, -1,7-, -1,8-, -2,3-, -2,4-, -2,5-, -2,6-, -2,7-, -2,8-,

- -3,4-, -3,5-, -3,6-, -3,7-, -3,8-, -4,5-, -4,6-, -4,7-, -4,8-, -5,6-, -5,7-, -5,8-, -6,7-, -6,8- or -7,8-diol,
- 2,7-diethyloctane-1,2-, -1,3-, -1,4-, -1,5-, -1,6-,

  5 -1,7-, -1,8-, -2,3-, -2,4-, -2,5-, -2,6-, -2,7-, -2,8-,

  -3,4-, -3,5-, -3,6-, -3,7-, -3,8-, -4,5-, -4,6-, -4,7-,

  -4,8-, -5,6-, -5,7-, -5,8-, -6,7-, -6,8- or -7,8-diol,
- 3,4-diethyloctane-1,2-, -1,3-, -1,4-, -1,5-, -1,6-,
  10 -1,7-, -1,8-, -2,3-, -2,4-, -2,5-, -2,6-, -2,7-, -2,8-,
  -3,4-, -3,5-, -3,6-, -3,7-, -3,8-, -4,5-, -4,6-, -4,7-,
  -4,8-, -5,6-, -5,7-, -5,8-, -6,7-, -6,8- or -7,8-diol,
- 3,5-diethyloctane-1,2-, -1,3-, -1,4-, -1,5-, -1,6-,

  -1,7-, -1,8-, -2,3-, -2,4-, -2,5-, -2,6-, -2,7-, -2,8-,

  -3,4-, -3,5-, -3,6-, -3,7-, -3,8-, -4,5-, -4,6-, -4,7-,

  -4,8-, -5,6-, -5,7-, -5,8-, -6,7-, -6,8- or -7,8-diol,
- 3,6-diethyloctane-1,2-, -1,3-, -1,4-, -1,5-, -1,6-,
  20 -1,7-, -1,8-, -2,3-, -2,4-, -2,5-, -2,6-, -2,7-, -2,8-,
  -3,4-, -3,5-, -3,6-, -3,7-, -3,8-, -4,5-, -4,6-, -4,7-,
  -4,8-, -5,6-, -5,7-, -5,8-, -6,7-, -6,8- or -7,8-diol,
  or
- 25 4,5-diethyloctane-1,2-, -1,3-, -1,4-, -1,5-, -1,6-, -1,7-, -1,8-, -2,3-, -2,4-, -2,5-, -2,6-, -2,7-, -2,8-, -3,4-, -3,5-, -3,6-, -3,7-, -3,8-, -4,5-, -4,6-, -4,7-, -4,8-, -5,6-, -5,7-, -5,8-, -6,7-, -6,8- or -7,8-diol.

The positionally isomeric diethyloctanediols for inventive use can be used as individual compounds or as mixtures of two or more diethyloctanediols for preparing the inventive oligomers and polymers.

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Particular advantages result from the use of 2,4-diethyloctane-1,5-diol.

The positionally isomeric diethyloctanediols for inventive use are compounds which are known per se and they can be prepared with the aid of customary and known synthesis methods of organic chemistry such as base-catalyzed aldol condensation or are obtained as by-products of large-scale chemical syntheses such as the preparation of 2-ethylhexanol.

Their fraction in the inventive oligomers and polymers may vary extremely widely and is guided by the particular end use of the inventive oligomers and polymers and by their profile of properties required for said use. In accordance with the invention it is of advantage if they are present in the starting products of the inventive oligomers and polymers in an amount, based on the starting products, of from 2 to 60% by weight, preferably from 3 to 55% by weight, with particular preference from 5 to 50% by weight, and in particular from 7 to 45% by weight.

The inventive oligomers and polymers come from the customary and known oligomer and polymer classes. For further details refer to Römpp Lexikon Lacke und Druckfarben, Georg Thieme Verlag, Stuttgart, New York, 1998, page 464, "Polymers". It is essential that, during the preparation of the inventive oligomers and polymers, the positionally isomeric diethyloctanediols for inventive use are incorporated into these oligomers and polymers as monomer units.

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The inventive oligomers and polymers are of linear or branched construction. In the inventive oligomers and polymers it is also possible to have combinations of linear chains with highly branched chains. In respect of the monomer units, the oligomer chains and polymer chains are of block, comb or random construction. Of course, these construction principles may be combined with one another as desired. Furthermore, the inventive oligomers and polymers may have a core/shell structure and/or may be present in the form of crosslinked microparticles.

Accordingly, the number-average molecular weight Mn and the molecular weight polydispersity Mw/Mn of the inventive oligomers and polymers may also vary extremely widely depending on end use and structure. In accordance with the invention, number-average molecular weights Mn of from 700 to 2 000 000 are of advantage. Within this range, further particular advantages are

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afforded by number-average molecular weights Mn of from 1000 to 1500000, in particular from 2000 to 1000000. The molecular weight polydispersity Mw/Mn in this case is advantageously from 1.1 to 20, preferably from 1.2 to 15 and in particular from 1.3 to 10.

Examples of advantageous inventive oligomers and are polyaddition resins, polycondensation polymers resins, or resins prepared by polyaddition and 10 polycondensation. For further details, refer to Römpp Lexikon Lacke und Druckfarben, Georg Thieme Verlag, Stuttgart, New York, 1998, page 457, "Polyaddition" and "Polyaddition resins (polyadducts)", and also pages 463 and 464, "Polycondensates", "Polycondensation", 15 "Polycondensation resins".

Examples of advantageous inventive polyaddition resins and/or polycondensation resins are polyethers, polyesters, polycarbonates, polyurethanes, polyureas, polyamides, polyimides or copolymers which contain ether, ester, carbonate, urethane, urea, amide and/or imide groups in the oligomer and/or polymer chains.

Examples of particularly advantageous inventive

25 polyaddition resins and/or polycondensation resins are
polyesters, polyurethanes, polyester-co-polyethers,
polyester-co-polycarbonates, polyester-copolyurethanes, polyester-co-polyamides, polyester-copolyureas or polyester-co-polyimides.

Of these, in turn, the polyesters, the polyurethanes, and the polyester-co-polyurethanes are especially advantageous.

- Viewed in terms of its method, their preparation has no special features but instead takes place in accordance with the customary and known methods of preparing polycondensation resins and polyaddition resins.
- 10 For example, the inventive polyesters are obtainable by reacting polycarboxylic acids and the above-described positionally isomeric diethyloctanediols for inventive and also, where appropriate, further polyols use, and/or monocarboxylic acids.

Examples of suitable polycarboxylic acids are aromatic polycarboxylic acids such as terephthalic isophthalic acid, phthalic acid, trimellitic acid or pyromellitic acid, and their ring-substituted alkyl

20 derivatives; cycloaliphatic polycarboxylic acids such as tetrahydrophthalic acid, 1,2-cyclobutanedicarboxylic acid, 1,3-cyclobutanedicarboxylic acid, 1,2-cyclopentanedicarboxylic acid, 1,3-cyclopentanedicarboxylic acid, hexahydrophthalic acid,

1,3-cyclohexane-

dicarboxylic acid, 1,4-cyclohexanedicarboxylic acid, 4-25 methylhexahydrophthalic acid ortricyclodecanedicarboxylic acid, which may be used both in their cis form and in their trans form and also as a mixture of both forms; or acyclic polycarboxylic acids such as

oxalic acid, malonic acid, succinic acid, glutaric acid, adipic acid, pimelic acid, suberic acid, azelaic acid, sebacic acid, undecanedicarboxylic acid or dodecanedicarboxylic acid, maleic acid, fumaric acid, itaconic acid, citraconic acid or dimer fatty acids.

Also suitable in accordance with the invention are the esterifiable derivatives of the abovementioned polycarboxylic acids such as their monoesters or polyesters with aliphatic alcohols having from 1 to 4 carbon atoms or hydroxy alcohols having from 1 to 4 carbon atoms, for example. It is also possible, moreover, to use the anhydrides of the abovementioned polycarboxylic acids, where they exist.

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Examples of suitable monocarboxylic acids that may be employed are 2-ethylhexanoic acid, 3,3,5-trimethylhexanoic acid, isononanoic acid, pelargonic acid, fatty acids from coconut oil, fatty acids of natural oils, resin acids, benzoic acid or p-tert-butylbenzoic acid.

Examples of suitable polyols, which may be used in addition to the positionally isomeric diethyl
25 octanediols for inventive use for the purposes of further varying the profile of properties, as starting products, are diols such as ethylene glycol, 1,2- or 1,3-propanediol, methylpropane-1,3-diol, ether oligomers of ethylene glycol and propylene glycol such

as diethylene glycol or dipropylene glycol, 1,2-, 1,3or 1,4-butanediol, 1,2-, 1,3-, 1,4- or 1,5-pentanediol, 1,2-, 1,3-, 1,4-, 1,5- or 1,6-hexanediol, neopentyl hydroxypivalate, neopentyl glycol, 1,2-, 1,3- or 1,4cyclohexanediol, 1,2-, 1,3or 1,4-cyclohexanedimethanol, tricyclodecanedimethanol (TCD), trimethylpentanediol, ethylbutylpropanediol, octanediols, nonanediols, 2-butyl-2-ethylpropane-1,3diol, 2-butyl-2-methylpropane-1,3-diol, 2-phenyl-2-10 methylpropane-1,3-diol, 2-propyl-2-ethylpropane-1,3diol. 2-di-tert-butylpropane-1,3-dio1, 2-butyl-2propylpropane-1,3-diol, 1-dihydroxymethylbicyclo[2.2.1]heptane, 2,2-diethylpropane-1,3-dio1, 2,2-dipropylpropane-1,3-diol 2-cyclohexyl-2-methyl-15 propane-1,3-diol, 2,5-dimethylhexane-2,5-diol, diethylhexane-2,5-diol, 2-ethyl-5-methylhexane-2,5diol, 2,4-dimethylpentane-2,4-diol, 2,3-dimethylbutane-2,3-dio1, 1,4-(2'-hydroxypropy1)-benzene, 1,3-(2'hydroxypropyl)-benzene or dimer diols from dimer fatty 20 acids; or triols such as glycerol, trimethylolethane, trimethylolpropane, trihydroxyethyl isocyanurate

The diols and the triols are employed here in a molar ratio which results in the desired degree of branching.

pentaerythritol.

The inventive polyesters are prepared by the known methods of esterification, such as are described, for example, in DE-A-40 24 204, page 4 lines 50 to 65. This

reaction normally takes place at temperatures between 180 and 280°C, if desired in the presence of a suitable esterification catalyst, such as lithium octoate, dibutyltin oxide, dibutyltin dilaurate or paratoluenesulfonic acid, for example.

The preparation of the inventive polyesters is normally conducted in the presence of small amounts of a suitable solvent as azeotrope former. Examples of azeotrope formers used include aromatic hydrocarbons, such as particularly xylene, and (cyclo)aliphatic hydrocarbons, e.g., cyclohexane or methylcyclohexane.

Where the inventive polyesters still contain at least one, preferably at least two, and in particular two, free hydroxyl groups, they may serve in a particularly advantageous way for preparing further inventive polymers, i.e., polyester-co-polymers, which constitutes a further very particular advantage of the inventive polyesters.

By way of example, the inventive polyurethanes are obtainable by the customary and known methods of polyurethane chemistry from polyisocyanates and the positionally isomeric diethyloctanediols for inventive use, and, if desired, further compounds which contain at least one, preferably at least two, functional groups which are reactive toward isocyanate groups. Instead of the positionally isomeric diethyloctanediols

for inventive use, or in addition to them, use is also made with particular advantage of the above-described inventive polyesters, thereby giving the inventive polyester-co-polyurethane.

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Examples of suitable polyisocyanates are isophorone diisocyanate (= 5-isocyanato-1-isocyanatomethyl-1,3,3trimethylcyclohexane), 5-isocyanato-1-(2-isocyanatoeth-1-y1)-1,3,3-trimethyl-cyclohexane, 5-isocyanato-1-(3isocyanatoprop-1-yl)-1,3,3-trimethyl-cyclohexane, isocyanato-(4-isocyanatobut-1-yl)-1,3,3-trimethyl-cyclohexane, 1-isocyanato-2-(3-isocyanatoprop-1-y1)cyclohexane, 1-isocyanato-2-(3-isocyanatoeth-1-1-isocyanato-2-(4-isocyanatobut-1-y1)yl)cyclohexane, cyclohexane, 1,2-diisocyanatocyclobutane, diisocyanatocyclobutane, 1,2-diisocyanatocyclopentane, 1,3-diisocyanatocyclopentane, 1,2-diisocyanatocyclohexane, 1,3-diisocyanatocyclohexane, 1,4diisocyanatocyclohexane, dicyclohexylmethane diisocyanate, liquid bis(4-isocyanatocyclohexyl)methane with a trans/trans content of up to 30% by weight, which is obtainable by phosgenating isomer mixtures of bis(4-aminocyclohexyl)methane or by fractional crystallization of commercial bis(4-isocyanatocyclohexyl) methane, as described in the patents DE-A-44 14 032, GB-A-1220717, DE-A-16 18 795 or DE-A-17 93 785, trimethylene diisocyanate, tetramethylene diisocyanate, pentamethylene diisocyanate, hexamethylene diisocyanate, ethylethylenediisocyanate,

trimethylhexane diisocyanate, heptanemethylene diisocyanate or diisocyanates derived from dimer fatty acids, as sold under the commercial designation DDI 1410 by Henkel and described in the patents WO 97/4974597/49747, especially 2-heptyl-3,4-bis(9-5 and isocyanatononyl)-1-pentylcyclohexane, or 1,2-, 1,4- or 1,3-bis(isocyanatomethyl)cyclohexane, 1,2-, 1,4- or 1,3-bis(2-isocyanatoethy-1-yl)cyclohexane, 1,3-bis(3isocyanatoprop-1-yl)cyclohexane, 1,2-, 1,4- or 1,3-10 bis(4-isocyanatobut-1-yl)cyclohexane, tolylene diisocyanate, xylylene diisocyanate, bisphenylene diisocyanate, naphthylene diisocyanate or diphenylmethane diisocyanate.

- Examples of suitable further compounds which may be employed as starting products and which contain at least one, preferably at least two, functional groups reactive toward isocyanate groups are
- 20 saturated and unsaturated high and low molecular mass diols and, in minor amounts, triols for introducing branches, especially the abovedescribed diols and triols,
- 25 polyamines,
  - amino alcohols, and

compounds by which stabilizing (potentially) ionic
 and/or nonionic functional groups are introduced.

Further examples of highly suitable compounds of this kind and also processes for preparing polyurethanes, especially polyester-co-polyurethanes, are known from the patents EP-A-0 073 517, EP-A-0 089 497, EP-A-0 228 003, EP-A-0 234 361, EP-A-0 543 817 or EP-A-0 574 417.

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Further particular advantages result if the inventive oligomers and polymers are reacted with olefinically unsaturated monomers to give graft copolymers.

- 15 Examples of suitable olefinically unsaturated monomers include
  - (meth)acrylic esters substantially free from acid groups,

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monomers which carry per molecule at least one hydroxyl group and are substantially free from acid groups, such as hydroxyalkyl esters of acrylic acid, methacrylic acid or another alpha, beta-olefinically unsaturated carboxylic acid which derive from an alkylene glycol which is esterified with the acid or which are obtainable by reacting the alpha, beta-olefinically

unsaturated carboxylic acid with an alkylene oxide,

- monomers which carry per molecule at least one
   acid group which can be converted into the corresponding acid anion group,
- vinyl esters of alpha-branched monocarboxylic acids having from 5 to 18 carbon atoms in the
   molecule,
- reaction products of acrylic acid and/or methacrylic acid with the glycidyl ester of an alpha-branched monocarboxylic acid having from 5 to 18 carbon atoms per molecule,
- cyclic and/or acyclic olefins such as ethylene,
   propylene, but-1-ene, pent-1-ene, hex-1-ene,
   cyclohexene, cyclopentene, norbonene, butadiene,
   isoprene, cylopentadiene and/or dicyclopentadiene,
   especially ethylene,
  - (meth)acrylamides,
- 25 monomers containing epoxide groups such as the glycidyl esters of ethylenically unsaturated carboxylic acids,
  - vinylaromatic hydrocarbons,

- nitriles,
- vinyl compounds, especially vinyl halides and/or vinylidene dihalides, N-vinylpyrrolidone, vinyl esters or vinyl ethers,
  - allyl compounds, especially allyl ethers and allyl esters.
- 10 Further examples of suitable olefinically unsaturated monomers and also suitable methods of graft copolymerization are described, for example, in the EP-A-0 521 928, patents EP-A-0 730 613 or EP-A-0 787 159.

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The inventive oligomers and polymers may functional groups which undergo crosslinking reactions with complementary functional groups. complementary functional groups may be present in the 20 inventive oligomers and polymers themselves, which as a result become self-crosslinking. Alternatively, complementary functional groups may be present compounds which exist separately from the inventive oligomers and polymers. Mixtures of this kind are 25 referred to by those in the art as externally crosslinking systems, and the relevant compounds as crosslinking agents. In these externally crosslinking systems, the inventive oligomers and polymers take over the function of the binders.

Examples of suitable complementary functional groups for inventive use which undergo crosslinking reactions are assembled in the following overview. In the overview the variable R stands for an acyclic or cyclic aliphatic radical, an aromatic radical and/or an aromatic-aliphatic (araliphatic) radical; the variables  $R^1$  and  $R^2$  stand for identical or different aliphatic radicals or are linked to one another to form an aliphatic or heteroaliphatic ring.

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# Overview: Examples of complementary functional groups in the

### Binder (self-crosslinking)

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# Binder and crosslinking agent

or

### Crosslinking agent and binder

20 -SH

-C(O)-OH

 $-NH_2$ 

-C(0)-O-C(0)-

-OH

-NCO

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 $-O-(CO)-NH-(CO)-NH_2$ 

-NH-C(O)-OR

 $-O-(CO)-NH_2$ 

-CH2-OH

 $-CH_2-O-CH_3$  $-NH-C(O)-CH(-C(O)OR)_2$ 5 -NH-C(O)-CH(-C(O)OR)(-C(O)-R) $-NH-C(O)-NR^1R^2$  $= Si(OR)_2$ 10 0  $-CH-CH_2$ 15 -C(O)-OH Ο  $-CH-CH_2$ -O-C(O)-CR=CH2 -OH20 -O-CR=CH<sub>2</sub>  $-NH_2$  $-C(0)-CH_2-C(0)-R$ 

Instead of these complementary functional groups or in addition to them, the inventive oligomers and polymers may contain functional groups which react with one

 $-CH=CH_2$ 

another or with other groups when exposed to actinic radiation. Examples of suitable groups of this kind are the allyl and vinyl groups set out above. Further examples of suitable groups are acrylate and methacrylate groups, especially acrylate groups. The actinic radiation may comprise electromagnetic radiation such as X-rays, UV radiation, visible light or near IR (NIR) light, or corpuscular radiation such as electron beams.

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The inventive oligomers and polymers described above are outstandingly suitable for preparing molding compounds, adhesives, and coating materials, especially paints.

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For preparing the molding compounds of the invention it is possible to use inventive liquid or solid oligomers and polymers which contain the functional groups described above, so that they can be processed conventionally by heating and/or by exposure to actinic radiation to give thermoset inventive moldings, films, and fibers. However, it is advantageous to use solid inventive oligomers and polymers which thermoplastic properties and no longer contain any reactive functional groups, so that they may be processed conventionally, for example, by extrusion followed by injection molding, film blowing or the drawing of fibers to give the thermoplastic inventive moldings, films, and fibers.

The inventive moldings, films, and fibers may be adhesively bonded outstandingly with the adhesives of the invention and/or coated or painted outstandingly with the inventive coating materials.

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The inventive molding compounds are composed of the inventive oligomers and polymers or they comprise conventional plastics additives in effective amounts.

10 For the preparation of inventive adhesives which serve as contact adhesives, it is preferred to use inventive oligomers and polymers which contain only a very small amount, if any, of the functional groups described above.

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For preparing inventive adhesives which serve as reactive adhesives, it is preferred to use inventive oligomers and polymers which possess a relatively large amount of the functional groups described above, so that they can be cured thermally and/or with actinic radiation. In addition, the inventive adhesives may comprise the additives described below for the coating materials, provided they are suitable for use in adhesives.

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By means of the inventive adhesives, the substrates described below, particularly the inventive moldings, films, and fibers, can be adhesively bonded outstandingly.

With particular verv preference, the inventive oligomers and polymers are used for preparing coating materials, especially paints. For this end use it is preferred to use oligomers and polymers which contain the above-described functional groups. The inventive coating materials here are self-crosslinking externally crosslinking in the sense referred to above, but especially externally crosslinking. Furthermore, they are curable thermally or with actinic radiation, or thermally and with actinic radiation, the latter being referred to by those in the art as dual cure.

The selection of the respective complementary functional groups is guided on the one hand by the consideration that during storage they must not undergo any unwanted reactions and/or, where appropriate, must not disrupt or inhibit the curing with actinic radiation, and secondly by the temperature range with which thermal curing is to take place.

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Particularly in respect of heat-sensitive substrates such as plastics, it is of advantage here in accordance with the invention to choose a temperature range which does not exceed 100°C, especially 80°C. In the light of these boundary conditions, hydroxyl groups and isocyanate groups or carboxyl groups and epoxy groups have proven advantageous as complementary functional groups, and so are employed preferably in accordance with the invention in the inventive coating materials

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that are in the form of two-component or multicomponent systems. Particular advantages result if the hydroxyl groups are used as functional groups in the binders and the isocyanate groups as functional groups in the crosslinking agents.

Where higher crosslinking temperatures, for example, from 100°C to 180°C, may be employed, inventive coating materials suitably include one-component systems, in functional groups which the in the binders are preferably thio, amino, hydroxyl, carbamate, allophanate, carboxyl and/or (meth)acrylate groups, but especially hydroxyl groups, and the functional groups in the crosslinking agents are preferably anhydride, carboxyl, epoxy, blocked isocyanate, urethane. methylol, methylol ether, siloxane, amino, hydroxyl and/or beta-hydroxyalkylamide groups.

In the context of the present invention, 20 component (1K) system is a thermosetting coating material in which the binder and the crosslinking agent are present alongside one another, i.e., in component. A prerequisite for this is that the two constituents crosslink with one another only 25 relatively high temperatures and/or on exposure to actinic radiation.

The inventive coating material may further be a two-component (2K) or multicomponent (3K, 4K) system.

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In the context of the present invention, this means a coating material in which, in particular, the binder and the crosslinking agent are present separately from one another in at least two components which are not combined until shortly before application. This form is chosen when binder and crosslinking agent react with one another even at room temperature. Coating materials of this kind are employed in particular for coating heat-sensitive substrates, especially in automotive refinish.

Not least, it is possible for the inventive coating materials to be physically curing, i.e., to crosslink by loss of solvent from the applied coating materials and/or by coalescence of the binder particles. Linking within the coatings takes place by looping of the polymer chains to the binders. For further details, refer to Römpp Lexikon Lacke und Druckfarben, Georg Thieme Verlag, Stuttgart, New York, 1998, pages 274 and 275, "Curing".

The inventive coating material is used for producing decorative and/or protective coatings, especially paint systems such as transparent clearcoats, primers, especially antistonechip primers and surfacers, or color and/or effect paint systems, especially topcoats and basecoats. These paint systems may comprise one or more coats.

For these end uses, it is normally the case that customary coatings additives in effective amounts are added to the inventive coating material. The nature and amount of the additives are guided in particular by the end use of the inventive coating material. Preferably, these additives are not volatile under the processing and application conditions of the inventive coating material.

10 Where the inventive coating material is used as a topcoat or basecoat material, it comprises color and/or effect pigments in customary and known amounts. The pigments may be composed of organic or inorganic compounds and may impart effect and/or color. Owing to this large number of suitable pigments, therefore, the inventive coating material ensures a universal scope for use of the coating materials and makes it possible to realize a large number of shades and optical effects.

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Furthermore, the inventive coating material may comprise organic and inorganic fillers in customary and known effective amounts. Examples of suitable fillers are chalk, calcium sulfates, barium sulfate, silicates such as talc or kaolin, silicas, oxides such as aluminum hydroxide or magnesium hydroxide, or organic fillers such as textile fibers, cellulose fibers, polyethylene fibers or wood flour.

These additives are omitted if the inventive coating materials are used as clearcoat materials.

Examples of suitable additives which may be present both in the clearcoat and in the topcoat materials of the invention are

- customary and known oligomeric and polymeric binders such as thermally curable hydroxylcontaining linear and/or branched and/or block, 10 comb and/or random poly(meth)acrylates or acrylate copolymers, polyesters, alkyds, aminoresins, polyurethanes, acrylated polyurethanes, acrylated polyesters, polylactones, polycarbonates, poly-15 ethers, epoxy resin-amine adducts, acrylatediols, partially hydrolyzed polyvinyl esters or polyureas, or actinic-radiation-curable (meth)acryloyl-functional (meth)acrylate copolymers, polyether acrylates, polyester 20 acrylates, unsaturated polyesters, epoxy acrylates, urethane acrylates, amino acrylates, melamine acrylates, silicone acrylates, and the corresponding methacrylates;
- 25 customary and known reactive diluents curable thermally and/or with actinic radiation, such as the diethyloctanediols per se for inventive use, (meth)acrylates with a functionality of two or

more, or polyisocyanates containing (meth)acrylate groups;

- UV absorbers and light stabilizers such as
   benzotriazoles, triazines or oxalanilides;
  - free-radical scavengers such as HALS compounds;
- thermolabile free-radical initiators such as organic peroxides, organic azo compounds or C-Ccleaving initiators such as dialkyl peroxides, peroxocarboxylic acids, peroxodicarbonates, peroxide esters, hydroperoxides, ketone peroxides, azo dinitriles or benzpinacol silyl ethers;

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- crosslinking catalysts such as dibutyltin dilaurate or lithium decanoate;
- type whose mechanism of action is based on an intramolecular variant of the hydrogen abstraction reactions such as occur diversely in photochemical reactions (by way of example, refer here to Römpp Chemie Lexikon, 9th expanded and revised edition,

  Georg Thieme Verlag Stuttgart, vol. 4, 1991) or cationic photoinitiators (by way of example, refer here to Römpp Lexikon Lacke und Druckfarben, Georg Thieme Verlag Stuttgart, 1998, pages 444 to 446),

especially benzophenones, benzoins or benzoin ethers, or phosphine oxides;

- slip additives;

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- polymerization inhibitors such as phosphites;
- defoamers;
- or aminimized alkanols and polyols, phenols and alkylphenols or anionic emulsifiers such as alkali metal salts or ammonium salts of alkanecarboxylic acids, alkanesulfonic acids, and sulfo acids of alkoxylated alkanols and polyols, phenols and alkylphenols;
- wetting agents such as siloxanes, fluorine compounds, carboxylic monoesters, phosphoric
   esters, polyacrylic acids and their copolymers, or polyurethanes;
  - adhesion promoters such as tricyclodecanedimethanol;

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- leveling agents;
- high-boiling organic solvents ("long solvents");

- film-forming auxiliaries such as cellulose derivatives;

- transparent fillers based on silica, alumina or
   zirconium oxide; for further details reference is
   made to Römpp Lexikon Lacke und Druckfarben, Georg
   Thieme Verlag, Stuttgart, 1998, pages 250 to 252;

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rheology control additives such as those known patents WO 94/22968, EP-A-0 276 501. EP-A-0 249 201 orWO 97/12945; crosslinked polymeric microparticles, such as disclosed, for 20 example, in EP-A-0 008 127; inorganic phyllosilicates such as aluminum-magnesium silicates, sodium-magnesium and sodium-magnesiumfluorine-lithium phyllosilicates of the montmorillonite type; silicas such as Aerosils; or 25 synthetic polymers containing ionic and/or associative groups such as polyvinyl alcohol, poly(meth)acrylamide, poly(meth)acrylic acid. polyvinylpyrrolidone, styrene-maleic anhydride or

ethylene-maleic anhydride copolymers and their

derivatives, or hydrophobically modified ethoxylated urethanes or polyacrylates;

- flame retardants and/or

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flatting agents.

Further examples of suitable coatings additives are described in the textbook "Lackadditive" [Additives for coatings] by Johan Bieleman, Wiley-WCH, Weinheim, New York, 1998.

These additives are added to the inventive coating materials in customary and known, effective amounts.

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The material composition of the inventive coating materials may vary extremely widely depending on their intended use, which is a further essential advantage of the coating materials. In general it is advisable to employ the amounts of binders, crosslinking agents, and additives that are known from the prior art. Of advantage in this context are inventive coating materials which [lacuna]

25 - from 1 to 90% by weight, preferably from 2 to 80% by weight, with particular preference from 3 to 75% by weight, and in particular from 4 to 70% by weight of at least one inventive oligomer and polymer,

from 0 to 60% by weight, preferably from 1 to 50% by weight, with particular preference from 2 to 45% by weight, and in particular from 4 to 40% by weight of at least one crosslinking agent, and

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- from 0 to 90% by weight, preferably from 0.1 to 80% by weight, with particular preference from 0.5 to 75% by weight, and in particular from 1 to 70% by weight of at least one additive,

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the percentages being based on the solids content of the inventive coating material concerned and adding up to 100% by weight.

15 The coating material may be present in different forms.

Thus, given an appropriate choice of its above-described constituents, it may be present in the form of a liquid coating material which is essentially free from organic solvents and/or water (100% system).

However, the coating material may also comprise a solution or dispersion of the above-described constituents in organic solvents and/or water. It is a further advantage of the coating material that in this case it is possible to establish solids contents of up to more than 80% by weight, based on the coating material.

Moreover, given an appropriate choice of its above-described constituents, the coating material may be a powder coating material. For this purpose, the crosslinking agent may have been microencapsulated if it is a polyisocyanate. This powder coating material may then, if desired, be dispersed in water, to give a powder slurry coating material.

The coating material may also be a two-component or multicomponent system described above in which at least the crosslinking agent is stored separately from the other constituents and is not added to them until shortly before use. In this case, the coating material of the invention may also be aqueous, the crosslinking agent preferably being present in a component comprising a solvent.

Furthermore, the coating material may be part of a so-called mixer system or modular system, as described, for example, in patents DE-A-41 10 520, EP-A-0 608 773, EP-A-0 614 951 or EP-A-0 471 972.

The preparation of the coating material from its constituents has no special features but instead takes place in a customary and known manner by mixing the constituents in appropriate mixing equipment such as stirred vessels, dissolvers or extruders in accordance with the techniques suitable for the preparation of the respective coating materials.

The coating material of the invention is used to produce the inventive coating systems, especially multicoat systems ML, on primed or unprimed substrates.

Suitable substrates are all surfaces to be coated which are not damaged by curing of the coatings present thereon using heat and/or actinic radiation; examples are the inventive moldings, films, and fibers; metals, ceramic, plastics, wood, stone, textile, 10 composites, leather, glass, glass fibers, glass wool, rock wool, mineral- and resin-bound building materials, such as plasterboard and cement slabs or roof shingles, and also assemblies of these materials. Accordingly, the multicoat system of the invention is also suitable 15 for applications outside of motor vehicle finishing, especially automobile finishing, being suitable particular for furniture coating and industrial coating, including coil coating and container coating. In the context of industrial coatings it is suitable 20 for coating virtually all parts, for private industrial use, such as radiators, domestic appliances, small metal parts, such as nuts and bolts, hubcaps, wheel rims, packaging or electrical components such as motor windings or transformer windings.

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In the case of electrically conductive substrates it is possible to use primers which are produced in a customary and known manner from electrodeposition coating materials. For this purpose, both anodic and

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cathodic electrodeposition coating materials are suitable, but especially cathodic materials.

Using the multicoat system of the invention it is also possible in particular to coat primed or unprimed plastics such as, for example, ABS, AMMA, ASA, CA, CAB, EP, UF, CF, MF, MPF, PF, PAN, PA, PE, HDPE, LDPE, LLDPE, UHMWPE, PET, PMMA, PP, PS, SB, PUR, PVC, RF, SAN, PBT, PPE, POM, PUR-RIM, SMC, BMC, PP-EPDM, and UP (abbreviated codes in accordance with DIN 7728T1). The plastics to be coated may of course also be polymer blends. modified plastics, or fiber reinforced plastics. It is also possible to employ the plastics commonly used in vehicle construction, especially motor vehicle construction.

Unfunctionalized and/or nonpolar substrate surfaces may be subjected prior to coating in a known manner to a pretreatment, such as with a plasma or by flaming, or may be provided with a water-based primer.

The multicoat systems ML of the invention may be produced in a variety of ways.

- 25 In a first preferred variant, the process of the invention comprises the following steps:
  - (I) preparing a surfacer film by applying a surfacer to the substrate,

- (II) curing the surfacer film to give the surfacer coat,
- (III) preparing a solid-color topcoat film by
  applying a solid-color topcoat material to the surfacer coat, and
  - (IV) curing the solid-color topcoat film to give the solid-color topcoat.

Another preferred variant of the process of the invention comprises the following steps:

- (I) preparing a basecoat film by applying a15 basecoat material to the substrate,
  - (II) drying the basecoat film,
- (III) preparing a clearcoat film by applying a clearcoat material to the basecoat film, and
  - (IV) jointly curing the basecoat film and the clearcoat film to give the basecoat and the clearcoat (wet-on-wet technique).

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A third preferred variant of the process of the invention comprises the steps of:

- (I) preparing a surfacer film by applying a surfacer to the substrate,
- (II) curing the surfacer film to give the surfacer
  5 coat,
  - (III) preparing a basecoat film by applying a basecoat material to the surfacer coat,
- 10 (IV) drying the basecoat film,
  - (V) preparing a clearcoat film by applying a clearcoat material to the basecoat film, and
- 15 (VI) jointly curing the basecoat film and the clearcoat film to give the basecoat and the clearcoat (wet-on-wet technique).
- Which of the preferred variants is chosen depends on the intended use of the multicoat systems ML of the invention. For instance, the third variant, in particular, is employed with especial preference in the context of automotive OEM finishing.
- 25 Accordingly, the multicoat systems ML of the invention may differ in their structure.

In a first preferred variant of the multicoat system ML of the invention,

- (1) a surfacer coat which absorbs mechanical energy, and
- (2) a color and/or effect topcoat

lie above one another in the stated sequence.

In a second preferred variant of the multicoat system ML of the invention,

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- (1) a surfacer coat which absorbs mechanical energy,
- (2) a color and/or effect basecoat, and
- 15 (3) a clearcoat

lie above one another in the stated sequence.

In a third preferred variant of the multicoat system ML of the invention,

- (1) a color and/or effect basecoat and
- (2) a clearcoat

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lie above one another in the stated sequence. The third preferred variant is employed in particular in the context of coating plastics.

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application of the coating material The of invention may take place by any customary application method, such as spraying, knife coating, brushing, flow coating, dipping, impregnating, trickling or rolling. The substrate to be coated may be at rest per se, with application equipment or unit being moved. Alternatively the substrate to be coated, particularly a coil, may be moved, with the application unit being at rest relative to the substrate or being moved appropriately.

It is preferred to employ spray application methods, such as compressed air spraying, airless spraying, high-speed rotation, electrostatic spray application (ESTA), for example, alone or in conjunction with hot spray application such as hot air spraying, for example. Application may take place at temperatures of max. 70 to 80°C, so that appropriate application viscosities are attained without any change or damage to the coating material and its overspray (which may be intended for reprocessing) during the short period of thermal stress. For instance, hot spraying may be configured in such a way that the coating material is heated only very briefly in the spray nozzle or shortly before the spray nozzle.

The spray booth used for application may, for example, be operated with a circulation system, which may be temperature-controllable, and which is operated with an

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appropriate absorption medium for the overspray, an example of such medium being the coating material itself.

5 Where the coating material includes constituents crosslinkable with actinic radiation, application is made under illumination with visible light with a wavelength of above 550 nm, or in the absence of light. By this means, material alteration or damage to the coating material and to the overspray is avoided.

In general, the surfacer film, topcoat film, basecoat film and clearcoat film are applied in a wet film thickness such that their curing results in coats having the thicknesses which are advantageous necessary for their functions. In the case of surfacer film, this thickness is from 10 to 150, preferably from 15 to 120, with particular preference from 20 to 100, and in particular from 25 to 90  $\mu$ m; in the case of the topcoat it is from 5 to 90, preferably from 10 to 80, with particular preference from 15 to 60, and in particular from 20 to 50  $\mu$ m; in the case of the basecoat it is from 5 to 50, preferably from 10 to 40, with particular preference from 12 to 30 and in particular from 15 to 25  $\mu m$ , and in the case of the clearcoats it is from 10 to 100, preferably from 15 to 80, with particular preference from 20 to 70, and in particular from 25 to 60  $\mu m$ .

In accordance with the invention, the surfacer film, topcoat film, basecoat film and clearcoat film may be cured thermally, with actinic radiation, or thermally and with actinic radiation, depending on their material composition. In accordance with the invention it is of advantage to cure the basecoat film only partly if at all before applying the clearcoat film, in order then to cure it together with the clearcoat film (wet-on-wet technique).

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Curing may take place after a certain rest period. This period may have a duration of from 30 s to 2 h, preferably from 1 min to 1 h, and in particular from 1 min to 30 min. The rest period is used, for example, for leveling and devolatilization of the coating films or for the evaporation of volatile constituents such as solvents, water or carbon dioxide if the coating material has been applied using supercritical carbon dioxide as solvent. The rest period may be shortened and/or assisted by the application of elevated temperatures up to 80°C, provided this does not entail any damage or alteration to the coating films, such as premature complete crosslinking, for instance.

25 The thermal curing has no special features in terms of its method but instead takes place in accordance with the customary and known methods such as heating in a forced-air oven or irradiation with IR lamps. Thermal curing may also take place in stages. Advantageously,

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it is effected at a temperature of from 50 to 100°C, with particular preference from 80 to 100°C, and in particular from 90 to 100°C, for a period of from 1 min to 2 h, with particular preference from 2 min to 1 h, and in particular from 3 min to 30 min. Where the substrates used have a high capacity to withstand thermal stress, thermal crosslinking may also be conducted at temperatures above 100°C. In general it is advisable in this case not to exceed temperatures of 180°C, preferably 160°C, and in particular 140°C.

Given appropriate material composition of the an coating material, the thermal curing may be supplemented by curing with actinic radiation, it being possible to use UV radiation and/or electron beams. If desired, it may be supplemented by or conducted with actinic radiation from other radiation sources. In the case of electron beams, it is preferred to operate under an inert gas atmosphere. This may be ensured, for example, by supplying carbon dioxide and/or nitrogen directly to the surface of the coating film.

In the case of curing with UV radiation, as well, it is possible to operate under inert gas in order to prevent the formation of ozone.

Curing with actinic radiation is carried out using the customary and known radiation sources and optical auxiliary measures. Examples of suitable radiation

sources are high or low pressure mercury vapor lamps, with or without lead doping in order to open up a radiation window of up to 405 nm, or electron beam sources. The arrangement of these sources is known in principle and may be adapted to the circumstances of the workpiece and the process parameters. In the case of workpieces of complex shape such as automobile bodies, the regions not accessible to direct radiation (shadow regions) such as cavities, folds and other structural undercuts may be cured using point, smallarea or all-round emitters, in conjunction with an automatic movement means for the irradiation of cavities or edges.

The equipment and conditions for these curing methods are described, for example, in R. Holmes, "U.V. and E.B. Curing Formulations for Printing Inks", Coatings and Paints, SITA Technology, Academic Press, London, United Kingdom, 1984.

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Curing here may take place in stages, i.e., by multiple exposure to light or actinic radiation. This may also be done alternately, i.e., by curing in alternation with UV radiation and electron beams.

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Where thermal curing and curing with actinic radiation are employed together (dual cure), these methods may be used simultaneously or in alternation. Where the two curing methods are used in alternation, it is possible,

for example, to begin with thermal curing and end with actinic radiation curing. In other cases it may prove advantageous to begin and to end with actinic radiation curing. The skilled worker is able to determine the curing method particularly appropriate to each individual case on the basis of his or her general knowledge in the art, possibly with the aid of simple preliminary experiments.

10 The multicoat systems ML of the invention have an outstanding profile of properties which is very well balanced in terms of mechanical properties, optical properties, corrosion resistance, and adhesion. Thus the multicoat systems ML of the invention have the 15 intercoat adhesion and high optical quality which the market requires and no longer give rise to any problems such as inadequate condensation resistance of surfacer coats, cracking (mud cracking) the basecoats, or leveling defects or surface structures in 20 the clearcoats.

In particular, the multicoat system ML of the invention has an outstanding metallic effect an outstanding D.O.I. (distinctiveness of the reflected image) and outstanding surface smoothness. It is stable to weathering, resistant to chemicals and bird droppings, is scratch resistant, and exhibits very good reflow properties.

A further important advantage is the very good overcoatability of the multicoat system ML of the invention, even without abrading. As a result, it can easily be coated with customary and known, highly scratch-resistant coating materials based on organically modified ceramic materials.

Not least, however, it proves to be a very particular advantage that by means of the processes described above it is possible to produce a multicoat system which is based exclusively on the inventive coating materials.

Accordingly, the substrates of the invention also have particular advantages such as a long service life, a better ethetic impression to the viewer, and better technological usefulness.

#### Examples

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#### Example 1

#### The preparation of the inventive polyester 1

25 A 4 liter stainless steel reactor equipped with stirrer, reflux condenser, water separator and regulatable heating was charged with 195 parts by weight of hexane-1,6-diol, 720 parts by weight of trimethylolpropane, 334 parts by weight of 2,4-

diethyloctane-1,5-diol, 131 parts weight of bу isononanoic acid, 1272 parts by weight of hexahydrophthalic anhydride and 104 parts by weight of xylene. The resulting mixture was slowly heated to not more than 225°C and at this temperature was condensed to an acid number of 16 mg KOH/g and a viscosity of in solvent naphtha). The 12 dPas (60% resulting reaction mixture was then allowed to cool and at 130°C was diluted with 1530 parts by weight of solvent naphtha. The resulting solution of the inventive polyester had a solids content of 58.5% by weight, an acid number of 16.3 mg KOH/g, and a viscosity of 11 dPas (original).

#### 15 **Example 2**

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#### The preparation of the inventive polyester 2

4 liter stainless steel reactor equipped with 20 stirrer, reflux condenser, water separator and regulatable heating was charged with 383 parts by weight of trimethylolpropane, 355 parts by weight of 2,4-diethyloctane-1,5-diol, 69 parts by weight of isononanoic acid, 676 parts by weight of 25 hexahydrophthalic anhydride and 59 parts by weight of xylene and the resulting mixture was slowly heated to not more than 225°C and at this temperature was condensed to an acid number of 17 mg KOH/g and a viscosity of 12 dPas (60% in solvent naphtha).

resulting reaction mixture was then allowed to cool and at 130°C was diluted with 840 parts by weight of solvent naphtha. The resulting solution of the inventive polyester had a solids content of 59.5% by weight, an acid number of 17.1 mg KOH/g, and a viscosity of 13 dPas (original).

#### Example 3

# 10 The preparation of the inventive clearcoat material 1

A stirred vessel with mechanical stirrer was charged with 68.6 parts by weight of the inventive polyester 1 from example 1, 1.0 parts by weight of Tinuvin® 292 (light stabilizer from Ciba-Geigy), 1.5 parts by weight 15 of Tinuvin<sup>®</sup> 1130 (light-stabilizer from Ciba-Geigy), 10 parts by weight of butyl diglycol acetate, 9 parts by weight of butyl glycol acetate, 7 parts by weight of methoxypropyl acetate, 9.3 parts by weight of butyl acetate and 0.2 parts by weight of BYK® 310 (leveling 20 agent from Byk), and these constituents were mixed. Shortly before the application of the resulting mixture, 24.1 parts by weight of a crosslinking agent based on hexamethylene diisocyanate were added, after 25 which the two-component system was homogenized.

# Example 4

The production of the inventive multicoat system ML 1

To produce the inventive multicoat system ML, use was made of customary and known steel test panels which had been coated with an electric coat, produced from a commercial electrocoat material, and with a surfacer coat, produced from a commercial surfacer. A commercial aqueous basecoat material from BASF Coatings AG was applied in a customary and known way to the surfacer coat, with a film thickness of 12 - 15 µm, and was flashed off at 80°C for 10 minutes. Finally, the inventive clearcoat material 1 from example 3 was applied with a film thickness of 40-45 µm, after which the basecoat and the clearcoat were cured at 140°C for 20 minutes (wet-on-wet technique).

- 15 The inventive multicoat system ML proved to be extremely resistant to condensation: constant condensation climate test to DIN 50017 and 53209: no blisters, index 0.
- 20 The intercoat adhesion was also outstanding: cross-cut with Tesa tearoff to DIN EN ISO 2409: GT 0.

The deformability of the coating to DIN EN ISO 1520: 1995-04 was likewise very good: Erichsen cupping: 6.2 mm.

These figures demonstrate the advantageous nature of the inventive multicoat system, of the inventive clearcoat material, and of the inventive polyester.

# Example 5

# The preparation of the inventive clearcoat material 2

5 Example 3 was repeated except that the inventive polyester 2 from example 2 was used instead of the inventive polyester 1 from example 1.

# Example 6

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## The production of the inventive multicoat system ML 2

Example 4 was repeated except that the inventive clearcoat material 2 from example 5 was used instead of the inventive clearcoat material 1 from example 3. The same advantageous results were obtained.

Oligomers and polymers based on diethyloctanediols, their preparation and use

#### Claims

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- 1. Oligomers and polymers which contain at least one positionally isomeric diethyloctanediol as monomer unit.
- 2. Oligomers and polymers as claimed in claim 1, characterized in that the positionally isomeric diethyloctanediol is a 2,3-, 2,4-, 2,5-, 2,6-, 2,7-, 3,4-, 3,5-, 3,6- or 4,5-diethyloctanediol.
- 15 3. Oligomers and polymers as claimed in claim 2, characterized in that the positionally isomeric diethyloctanediol is a 2,4-diethyloctanediol.
- 4. Oligomers and polymers as claimed in any of claims

  1 to 3, characterized in that the positionally isomeric diethyloctanediol is a diethyloctane-1,2-, -1,3-, -1,4-, -1,5-, -1,6-, -1,7-, -1,8-, -2,3-, -2,4-, -2,5-, -2,6-, -2,7-, -2,8-, -3,4-, -3,5-, -3,6-, -3,7-, -3,8-, -4,5-, -4,6-, -4,7-, -4,8-, -5,6-, -5,7-, -5,8-, -6,7-, -6,8- or -7,8-diol.

5. Oligomers and polymers as claimed in claim 4, characterized in that the positionally isomeric diethyloctanediol is a diethyloctane-1,5-diol.

- 5 6. Oligomers and polymers as claimed in claim 5, characterized in that the positionally isomeric diethyloctanediol is a 2,4-diethyloctane-1,5-diol.
- Oligomers and polymers as claimed in any of claims
   1 to 6, characterized in that the resins in question
   are polyaddition resins and/or polycondensation resins.
- 8. Oligomers and polymers as claimed in claim 7, characterized in that the polyaddition resins and/or polycondensation resins are polyethers, polyesters, polycarbonates, polyurethanes, polyureas, polyamides, polyimides or copolymers which contain ether, ester, carbonate, urethane, urea, amide and/or imide groups in the oligomer and/or polymer chains.

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- 9. Oligomers and polymers as claimed in claim 8, characterized in that the polyaddition resins and/or polycondensation resins are polyesters, polyurethanes, polyester-co-polyethers, polyester-co-polycarbonates, polyester-co-polyurethanes, polyester-co-polyurethanes, polyester-co-polyurethanes, polyester-co-polyureas or polyester-co-polyimides.
- 10. Oligomers and polymers as claimed in any of claims 1 to 9, characterized in that they are of linear and/or

branched and/or block, comb and/or random construction, have a core/shell structure and/or are in the form of crosslinked microparticles.

- 5 11. Oligomers and polymers as claimed in any of claims 1 to 10, characterized in that they contain
- functional groups which undergo crosslinking reactions with complementary functional groups
   present in the oligomers and polymers themselves and/or in separate compounds, and/or
- functional groups which on exposure to actinic radiation react with one another or with other
   groups.
  - 12. Oligomers and polymers as claimed in any of claims 1 to 12, characterized in that they are grafted with olefinically unsaturated monomers.
  - 13. Use of the oligomers and polymers as claimed in any of claims 1 to 12 for preparing molding compounds, adhesives, and coating materials, especially paints.
- 25 14. Molding compounds, adhesives, and coating materials, characterized in that they comprise at least one oligomer and/or polymer as claimed in any of claims 1 to 12.

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15. Use of the molding compounds, adhesives, and coating materials as claimed in claim 14 for producing moldings, films, fibers, adhesive films, and decorative and/or protective coatings, especially paint systems.

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- 16. Moldings, films, and fibers producible from molding compounds as claimed in claim 14.
- 17. Adhesive films producible from adhesives as 10 claimed in claim 14.
  - 18. Coatings, especially paint systems, producible from coating materials, especially paints, as claimed in claim 14.

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19. Substrates which comprise at least one adhesive film as claimed in claim 17 and/or at least one coating, especially paint system, as claimed in claim 18.

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20. Substrates as claimed in claim 19, characterized in that they are moldings, films, and fibers as claimed in claim 16, motor vehicle bodies, industrial components, including electrical components, coils, and packaging, or furniture.

#### **ABSTRACT**

The invention relates to oligomers and polymers that contain at least one positional isomer diethyl octanediol as a monomer unit. The invention also relates to a method for producing the same and to the use thereof for producing molding compounds, adhesives and coating materials, especially lacquers. The invention especially relates to polyesters on the basis of 2,4-diethyloctane-1,5-diol and the use thereof in 2k-polyurethane lacquers.

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	DECLARATION FOR UTILITY OR	Attorney Docket No.	IN - 5554
ş	DESIGN	First Named Inventor	Hubert BAUMGART et al.
	PATENT APPLICATION (37 CFR 1.63)	СОМР	LETE IF KNOWN

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with initial Filin	g (surcharge	Filing Date	1	Wednesday, Fe	bruary 13, 2002
Filing (37	CFR 1.16 (e))	Group Art Unit			
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As below named inventor, I hereby declare the	at:				
My residence, post office address and citizen	ship are as stated below	next to my name.			
I believe I am the original, first and sole inven	tor (if only name is listed	d below) or an origina	al, first and	joint inventor (if pl	lural names are
listed below) of the subject matter which is cla					THOD FOR
OLIGOMERS AND POLYMER		OF DIETHYL-C	JCTAN.	EDIOLS, ME	HODFOR
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x is attached hereto					
x Was filed on 14. August 2000	an United States Ar	oplication or PCT Inte	rnational A	Application Numbe	. DCT/ED00/07900
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I hereby state that I have reviewed and under by any amendment specifically referred to ab		e above identified sp	ecification,	, including the clair	ms, as amended
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listed below and have also identified below, b	y checking the box, any	foreign application(s	<ul><li>s) for pater</li></ul>	nt or inventor's cer	tificate, or any PCT
international application(s) having a filing date	before that of the appli	cation on which priori	ity is claim	ed.	
Prior Foreign Application Country Number(s)	Foreign Filing   DATE/MONTH/			Yes	opy Attached? No
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Additional foreign application n	umber are listed on a su	pplemental priority da	ata sheet F	PTO/SB/02B attac	hed hereto:
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numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto

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Title Line Two::	DIETHYL-OCTANEDIOLS, METHOD FOR
Title Line Three::	PRODUCING SAME AND THE USE THEREOF
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Foreign Application One::	199 38 758.3
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Country::	Germany
Priority Claimed::	Yes

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